

Elevated ozone in the troposphere over the Atlantic and Pacific oceans in the Northern Hemisphere

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[1] Tropospheric column ozone (TCO) is derived from differential measurements of total column ozone from Total Ozone Mapping Spectrometer (TOMS), and stratospheric column ozone (SCO) from the Microwave Limb Sounder (MLS) instrument on the Upper Atmosphere Research Satellite (UARS). It is shown that TCO during late spring and summer months over the Atlantic and Pacific oceans at northern mid-latitudes is about 50–60 Dobson Units (DU) which is about the same as over the continents of North America, Europe and Asia (except high altitude mountain regions), where surface emissions of NO_x from industrial sources, biomass and biofuel burning, and biogenic emissions are significantly larger. The zonal characteristics of TCO derived from satellite measurements are generally simulated by a global chemical transport model called MOZART-2, but some discrepancies are also shown. The model results are analyzed to delineate the relative importance of surface NO_x emission, lightning NO_x and stratospheric flux. **INDEX TERMS:** 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3362 Meteorology and Atmospheric Dynamics: Stratosphere/troposphere interactions; 3367 Meteorology and Atmospheric Dynamics: Theoretical modeling. **Citation:** Chandra, S., J. R. Ziemke, X. Tie, and G. Brasseur (2004), Elevated ozone in the troposphere over the Atlantic and Pacific oceans in the Northern Hemisphere, *Geophys. Res. Lett.*, 31, L23102, doi:10.1029/2004GL020821.

1. Introduction

[2] Ozone is a precursor molecule of the hydroxyl (OH) radical which is the main oxidizing agent of several pollutants in the troposphere. In the troposphere ozone is produced primarily by photochemical oxidation of hydrocarbons in the presence of NO_x (NO + NO₂) with additional contribution from the stratosphere through stratosphere–troposphere exchange (STE). It is generally believed that tropospheric ozone has been increasing since pre-industrial times as a result of increased concentration of ozone-producing pollutants in Europe and North America [e.g., *Lelieveld and Dentener, 2000; Hauglustaine and Brasseur,*

2001; *Lelieveld et al., 2002*]. There is concern that with industrialization of Asian countries tropospheric ozone may be increasing in the Northern Hemisphere (NH) through long-range transport. Global models of chemistry and transport have been used to assess the contribution of Asian pollution over regions of Asia, North America, and Europe [e.g., *Berntsen et al., 1999; Li et al., 2001; Liu et al., 2002; Phadnis et al., 2002*]. Model results are usually compared with ozonesonde measurements which are few and far between. At NH mid-latitudes, column ozone derived from ozonesonde measurements tends to peak during summer months when anthropogenic emissions resulting from fossil fuel combustion and biomass burning are high [*Logan, 1999*]. It is difficult to assess the global implications of these results particularly over the vast regions of the Atlantic and Pacific oceans where ozone measurements are sparse. Comparisons of satellite measurements of TCO with global models have been limited mostly to tropical regions because of lack of satellite measurements of TCO outside the tropics.

[3] The purpose of this paper is to use TCO data from TOMS/MLS [*Chandra et al., 2003*] to characterize the zonal properties of TCO at NH mid-latitudes and to study the implications of various processes affecting TCO by using a global 3-D chemical transport model called MOZART version 2 [*Horowitz et al., 2003*]. The zonal and seasonal characteristics of TOMS/MLS TCO between ±30° was analyzed in detail by *Chandra et al. [2003]* and compared with a global 3-D model of tropospheric chemistry (GEOS-CHEM) for 1996–1997. In this paper a similar comparison of TOMS/MLS TCO is made with the MOZART-2 model to delineate the relative importance of STE, lightning, and anthropogenic NO_x emission. As *Chandra et al. [2003]* showed, TCO is derived using version 7 TOMS measurements with reflectivity <0.2. In addition, the calibration of MLS is adjusted to TOMS by normalizing MLS SCO to TOMS SCO derived from the convective cloud differential method.

2. TCO From TOMS/MLS

[4] TOMS/MLS measurements overlap for about 20 months (September 1991–April 1993) during the Nimbus-7 TOMS lifetime and for about 2 years (August 1996 to mid-1998) during the Earth Probe (EP) TOMS period. The frequency of MLS measurements also changes from almost daily measurements during the Nimbus-7 period to only a few days per month (5–10 days) during the EP TOMS period. The MLS measurements outside ±34° are available around every alternate month on average because of a 57° inclination of the UARS orbit and planned rotation of the satellite through yaw about every 36 days. Because the MLS instrument does not measure ozone below 100 hPa, zonal maps of TCO are most reliable between ±30°

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